

Phase coherence of conduction electrons below the Kondo temperature

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We have measured the phase decoherence rate, τ_ϕ^{-1} of conduction electrons in disordered Ag wires implanted with 2 and 10 parts per million Fe impurities, by means of the weak localization magnetoresistance. The Kondo temperature of Fe in Ag, $T_K \approx 4$ K, is in the ideal temperature range to study the progressive screening of the Fe spins as the temperature T falls below T_K . The contribution to τ_ϕ^{-1} from the Fe impurities is clearly visible over the temperature range 40 mK – 10 K. Below T_K , τ_ϕ^{-1} falls rapidly until $T/T_K \approx 0.1$, in agreement with recent theoretical calculations. At lower T , τ_ϕ^{-1} deviates from theory with a flatter T -dependence. We speculate that this latter behavior is due to incomplete screening of the $s=2$ Fe impurities by the conduction electrons.

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The Kondo problem is a paradigm many-body problem in condensed matter physics. In recent years, Kondo physics has been observed in semiconducting quantum dots [1] and carbon nanotubes, while Kondo lattices play a major role in some strongly-correlated materials [2]. The original problem that motivated Kondo's 1964 paper [3] was the increase in resistivity at low temperature of metals containing magnetic impurities. Although Kondo solved this puzzle, his perturbation theory diverged in the limit of zero temperature. Wilson [4] showed that, at temperatures far below the Kondo temperature T_K , the magnetic impurity forms a spin-singlet with the surrounding conduction electrons and behaves as a non-magnetic scatterer with cross section given by the unitarity limit.

The brief history given above might leave the impression that the behavior of dilute magnetic impurities in metals is completely understood. That is, however, not the case. An important aspect of the Kondo problem concerns the distinction between elastic and inelastic scattering. This distinction is extremely important in the context of quantum transport and mesoscopic physics, where it is known since 1979 that elastic scattering from static disorder preserves quantum phase coherence, whereas inelastic scattering destroys it. The magnetic impurity contribution to the conduction electron phase coherence rate, τ_ϕ^{-1} , was first measured explicitly by two groups in 1987 [5, 6], and has received renewed attention recently [7, 8] in the context of the debate over zero-temperature decoherence in disordered metals [9, 10]. Until very recently [11, 12], however, there was no theoretical expression for the temperature dependence of the inelastic scattering rate valid for T not too far below T_K , and very little reliable data in that temperature range [13].

The most reliable estimates of τ_ϕ^{-1} come from analysis of low-field magnetoresistance data in the context of weak-localization theory. In disordered metals without magnetic impurities, τ_ϕ^{-1} is dominated by electron-phonon scattering at temperatures above about 1 K, and by electron-electron scattering at lower T . In the pres-

ence of magnetic impurities, τ_ϕ^{-1} contains the additional contribution γ_m , which peaks at $T \approx T_K$. In order to observe this peak in γ_m while keeping the magnetic impurity concentration low enough to avoid interactions between impurities, one must choose a system with T_K below about 10 K; otherwise τ_ϕ^{-1} is dominated by electron-phonon scattering. In order to acquire data far below T_K , however, it is important to keep T_K as high as possible. The optimal range for T_K is a few Kelvins, which is achieved with Fe impurities in Ag [14].

We fabricated Ag wires of dimensions $L = 780$ μm , $w = 0.1 - 0.2$ μm and $t = 47$ nm on oxidized Si substrates using electron-beam lithography, thermal evaporation from a 99.9999%-purity Ag source, followed by lift-off of the bilayer resist. All the wires studied were evaporated simultaneously to ensure similar microstructure and resistivity. The 15 samples were divided into four batches – one that was kept pure, and three that were implanted with 2, 6, and 10 ppm of Fe impurities, respectively. SRIM simulations of the implantation at 70 keV indicate that over 90% of the implanted ions stay in the Ag wires, with the rest going into the substrate [15]. As none of the 6 ppm samples survived, we report data taken on a pure sample, and samples with 2 and 10 ppm Fe impurities. Table I lists the sample parameters. All samples were measured immersed in the mixing chamber of a dilution refrigerator with filtered leads. Four-probe resistance measurements were made using a lock-in amplifier with a ratio transformer to improve sensitivity [10]. The voltage drop across the sample was limited to $eV_s \lesssim k_B T$ to avoid heating the electrons.

Raw magnetoresistance (MR) data for the three samples, at $T = 1.8$ K, are shown in the inset to Fig. 1. MR data for each sample were fit using the following procedure. Because Ag has moderate spin-orbit scattering ($\tau_{so} \approx 40$ ps for all of our samples), the MR is positive at low temperatures, when $\tau_\phi \gg \tau_{so}$. At higher temperatures, when $\tau_\phi < \tau_{so}$, the MR starts out positive but then turns around at a field scale $B \approx 20$ mT. Data at several temperatures in this higher temperature range

Sample	L (μm)	t (nm)	w (nm)	R (Ω)	D (cm^2/s)	L_{so} (μm)	L_{ϕ}^{max} (μm)	c_{imp} (ppm)
Ag	780	47	130	3307	146	0.76	14.6	-
AgFe1	780	47	110	3890	146	0.86	9.1	2
AgFe2	780	47	185	2330	146	0.72	4.1	10

TABLE I: Geometrical and electrical characteristics of the measured samples at 1.2 K. L , t , and w are the sample dimensions and R is the resistance. D is the diffusion constant determined from the resistivity and the Einstein relation $\rho^{-1} = e^2 D \nu_F$, with the density of states in Ag $\nu_F = 1.03 \times 10^{47} \text{J}^{-1} \text{m}^{-3}$. L_{so} is the spin orbit length extracted from the fits of the magnetoresistance to weak localization theory. L_{ϕ}^{max} is the maximum coherence length measured at 40 mK. c_{imp} is the implanted Fe concentration.

(which was different for each sample) were first fit with three free parameters: $L_{\phi} = (D\tau_{\phi})^{1/2}$, where D is the diffusion constant, $L_{so} = (D\tau_{so})^{1/2}$, and the sample width, w . For each sample, these fits gave consistent values of L_{so} and w over a broad temperature range [16]. We then fixed those values of L_{so} and w , and fit the MR curves for all temperatures with L_{ϕ} as the only free parameter. Using the value of D obtained from the resistance and sample dimensions, we finally obtain τ_{ϕ} as a function of temperature for each sample.

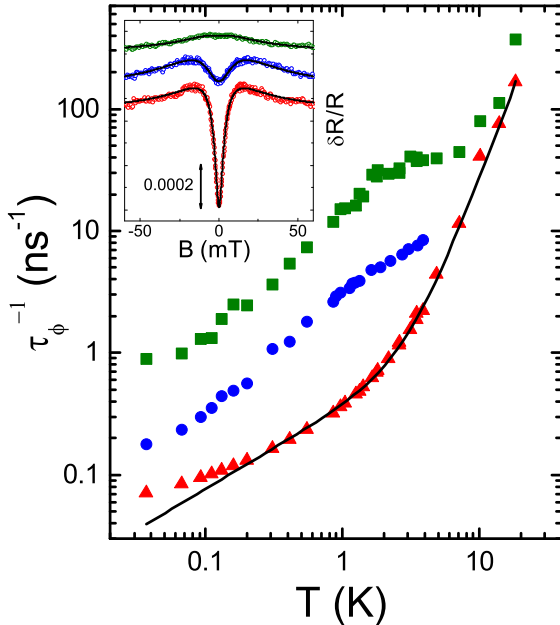


FIG. 1: (color online) Total decoherence rate vs. temperature for a pure Ag sample (\blacktriangle), and for samples with implanted Fe concentrations of 2 ppm (\bullet) and 10 ppm (\blacksquare). Inset: Raw magnetoresistance data at $T=1.8$ K for the three samples, and fits to weak localization theory.

Fig. 1 shows the decoherence rate, τ_{ϕ}^{-1} , obtained from the MR data and fitting procedure described above, for the three samples. The τ_{ϕ}^{-1} data for the pure sample follow theoretical expectations above 200 mK, with a modest amount of saturation at lower temperature [10]. Above 200 mK, the data are fit well (solid line) by a combination of electron-phonon scattering ($\tau_{\phi}^{-1} \propto T^3$) and electron-electron scattering ($\tau_{\phi}^{-1} \propto T^{2/3}$). The prefactor of the $T^{2/3}$ term is equal to $0.36 \text{ ns}^{-1} \text{K}^{-2/3}$, in good agreement with the theoretical value [17]. The additional contribution to τ_{ϕ}^{-1} from the Fe impurities, γ_m , is clearly observable in the 2 ppm sample up to 4 K and in the 10 ppm sample up to 10 K. Above 10 K extraction of γ_m from τ_{ϕ}^{-1} is not reliable, due to an apparent difference in the magnitude of the electron-phonon scattering rate in the 10 ppm and pure samples.

The suppression of the weak localization magnetoresistance by magnetic impurities was first discussed in [18] for the case of static impurity spins. Later, Fal'ko [19] pointed out that the impurity spins have their own internal dynamics given by the Korringa rate, $\gamma_K \propto k_B T$. Since spin-flip scattering of conduction electrons and re-orientation of the impurity spins arise from the same processes, the ratio of their rates is given by $\gamma_m/\gamma_K = n_s/\nu_F k_B T$, where n_s is the density of magnetic impurities and ν_F is the total density of states of conduction electrons at the Fermi level. Below T_K the Korringa rate saturates, so the ratio becomes $\gamma_m/\gamma_K = n_s/\nu_F k_B T_K$. If $\gamma_K > \gamma_m$, then the impurity spin configuration is randomized during the time between spin-flip scattering events of the conduction electrons. The contribution of magnetic scattering to the total decoherence rate in the spin-singlet channel of the weak localization magnetoresistance is given by:

$$\tau_{\phi}^{-1} = \tau_{in}^{-1} + 2\gamma_m \quad \text{for} \quad \gamma_m > \gamma_K. \quad (1)$$

$$\tau_{\phi}^{-1} = \tau_{in}^{-1} + \gamma_m \quad \text{for} \quad \gamma_m < \gamma_K. \quad (2)$$

where τ_{in}^{-1} is the dephasing rate due to electron-electron and electron-phonon scattering. For our Ag sample with 2 ppm Fe, assuming T_K in the range 2-4 K (see below), we find $\gamma_m/\gamma_K = 0.02 - 0.04 \ll 1$, hence we should use Eq. (2) to extract γ_m from τ_{ϕ}^{-1} . Fig. 2 shows γ_m for the 2 ppm and 10 ppm samples obtained from the τ_{ϕ}^{-1} data in Fig. 1 using Eq. (2), with τ_{in}^{-1} obtained from the data on the nominally pure sample. The data sets from the 2 ppm and 10 ppm samples are consistent with each other when we use Eq. (2) to analyze both of them.

The criterion $\gamma_m/\gamma_K \ll 1$ is a necessary condition for the theoretical approach of Micklitz *et al.* [12] in their numerical renormalization group (NRG) calculations of γ_m . Those authors note, however, that Eq. (2) is not strictly correct, because the decoherence induced by electron-electron interactions [17] is not an exponential process.

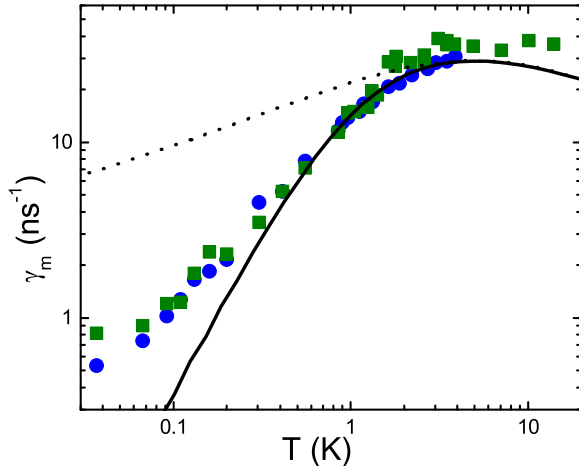


FIG. 2: (color online) Inelastic scattering rate due to magnetic impurities for the 2 ppm (●) and 10 ppm (■) samples. Data for the 2 ppm sample are multiplied by 5. The solid line is the theoretical calculation of Micklitz *et al.* [12] fit to the data for $T > 0.4$ K. The dotted line is the Suhl-Nagaoka approximation for $s=1/2$.

Eq. (12) in [12] shows how to add the decoherence rate due to e-e interactions with the rate due to all other processes including γ_m . We have compared the results of analyzing our data with Eq. (2) and with Eq. (12) in [12], and we find that for the 10 ppm sample the difference is negligible. For the 2 ppm sample, using Eq. (12) in [12] would increase γ_m by at most 15% at the lowest temperature, which is not significant on the logarithmic plot in Fig. 2. The solid line in Fig. 2 shows a fit of the NRG calculation from [12] to our data at temperatures above 0.4 K only. Fitting to the 2 ppm (10 ppm) data alone gives $T_K = 4.8$ K (5.4 K). For comparison, the Suhl-Nagaoka approximation with $T_K = 4.8$ K is shown as the dotted line. The data and fits shown in Fig. 2 lead immediately to several striking conclusions. First, as noted recently [13], the Suhl-Nagaoka approximation does not come close to reproducing the temperature dependence of γ_m for $T < T_K$. Second, the NRG theory of Micklitz *et al.* [12] fits the data reasonably well over the temperature range $T/T_K = 0.1 - 2$. And third, the NRG theory deviates strongly from the data when $T/T_K < 0.1$.

According to [12], the maximum value of γ_m , occurring at $T = T_K$, is $0.23 \times 4n_s/(\pi\hbar\nu_F)$. For 10 ppm of magnetic impurities in Ag, the theoretical estimate is $\tau_\phi^{max} = 16$ ns⁻¹, whereas the data in Fig. 2 show a maximum value about twice as large. This significant discrepancy may point to the inadequacy of the spin-1/2 theory of [12] to account for the large spin ($s=2$) of Fe impurities in Ag. On the other hand, previous measurements of γ_m in Ag samples implanted with Mn impurities [10], with $s=5/2$ and $T_K \approx 40$ mK, were consistent with

the theoretical estimate. (Those data were analyzed with the Suhl-Nagaoka approximation, which is in close agreement with the theory of [12] for $T > T_K$.) It thus appears that the inelastic scattering cross section of Fe in Ag is roughly twice that of Mn in Ag.

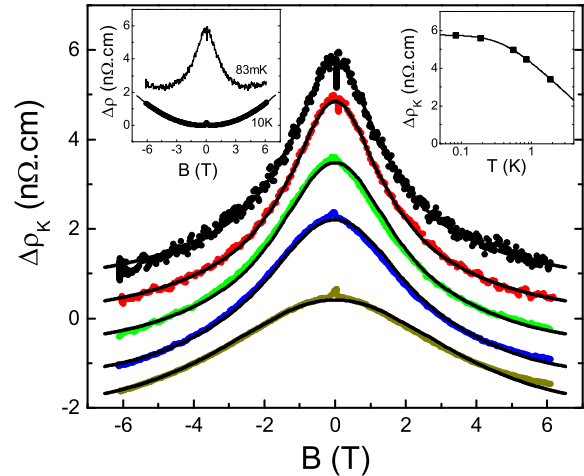


FIG. 3: (color online) Change in resistivity vs. magnetic field for the 10 ppm sample at temperatures of 83, 188, 550, 860, and 1880 mK, from top to bottom. The zero for the 83 mK data is chosen so that the fit curve approaches $\Delta\rho \rightarrow 0$ for $|B| \gg B_K$. Subsequent data sets are shifted downward by 0.75 nΩcm for clarity. Solid lines represent the global fit to the spin-1/2 NRG theory from [20], as discussed in the text. Left inset: Raw $\Delta\rho(B)$ data at 83 mK and 10 K. The latter shows the classical magnetoresistance $\Delta\rho(B) \propto B^2$, which was subtracted from the raw data sets to produce the curves in the main figure. Right inset: $\Delta\rho(B=0)$ vs. temperature.

To further test the theory, we seek an independent estimate of T_K to compare with the value obtained from the fit to γ_m shown in Fig. 2. The traditional method of determining T_K from the resistivity vs. temperature, $\rho(T)$, is not reliable for our samples, because the electron-phonon scattering contribution to $\rho(T)$ starts to grow significantly when $T > 8$ K, before the Kondo contribution has completely died out. Instead, we analyze the high-field magnetoresistivity, $\rho(B)$, shown at several temperatures in Fig. 3. $\rho(B)$ data for $T = 53$ mK (not shown) are indistinguishable from those at 83 mK, indicating that the Kondo contribution to ρ is close to the unitarity limit at those temperatures. We fit the $\rho(B)$ data using NRG calculations from ref. [20]. For $T \ll T_K$, these are nearly identical to the $T = 0$ analytical solution from [21]. The two-parameter fit to the 83 mK data set gives the values $\Delta\rho_K \equiv \rho(B=0, T=0) - \rho(B \gg B_K, T=0) = 0.57$ nΩcm/ppm and $B_K = 1.2$ T, where $g\mu_B B_K = k_B T_K$, with μ_B the Bohr magneton [22]. Using $g=2$, that in turn gives $T_K = 1.6$ K. Similar fits to the $\rho(B)$ data sets at higher temperatures, keeping $\Delta\rho_K$

fixed, show that they can not be fit consistently with such a low value of T_K .

To circumvent this problem, we have performed a global fit to all the $\rho(B)$ data sets with only three parameters: $\Delta\rho_K$, B_K , and T_K , where B_K determines the magnetic field scale over which $\rho(B)$ decreases, while T_K determines the temperature scale over which $\rho(B=0)$ decreases. The results of this global fit are shown by the solid lines in Fig. 3, with the values $B_K = 1.36$ T, $T_K = 2.96$ K, and $\Delta\rho_K = 0.58$ n Ω cm/ppm. The value of B_K corresponds to a Kondo temperature of 1.8 K. The discrepancy between the values of T_K extracted from the field scale and from the temperature scale highlights the fact that we are using a spin-1/2 theory. For free spins in a metal without Kondo effect, the magnetoresistance is proportional to $-\langle s_z \rangle^2$, where $\langle s_z \rangle$ is the average spin polarization given by the appropriate Brillouin function. A comparison of the functions $-\langle s_z \rangle^2$ for $s=1/2$ and $s=2$ shows that the full width at half maximum of the $s=1/2$ function is 1.85 times larger than for the $s=2$ function. If a similar relationship holds for the $\rho(B)$ curves in the Kondo regime, then the value $B_K = 1.2$ T we found from the $s=1/2$ fit to the 83 mK data would become $B_K \approx 2.2$ T for $s=2$, which corresponds to a Kondo temperature $T_K = 3.0$ K. That is consistent with the value of T_K obtained from the temperature dependence of $\rho(B=0)$, but still lower than the value $T_K \approx 5$ K obtained from the fit to γ_m .

The experimental value $\Delta\rho_K = 0.58$ n Ω cm/ppm should be compared with the unitarity limit for s-wave scattering, $\Delta\rho_K = 4\pi\hbar n_s/ne^2 k_F = 0.43$ n Ω cm/ppm for Ag, where k_F is the Fermi wavevector. (The value for d-wave scattering is five times higher.) Our measured value of $\Delta\rho_K$ is larger than the theoretical s-wave value, but less than the measured values in other Fe-noble metal Kondo systems: 1.3 n Ω cm/ppm in Cu:Fe and 1.0 n Ω cm/ppm in Au:Fe [23].

The most serious discrepancy between theory and experiment is the flattening of $\gamma_m(T)$ for $T/T_K < 0.1$, shown in Fig. 2. The behavior of the data is reminiscent of Eq. (3b) in [24], which describes inelastic scattering by an underscreened Kondo impurity when $T \ll T_K$. Since the magnitude of γ_m at $T = 40$ mK is 40 times smaller than at 4 K, however, fitting the low- T data with Eq. (3b) of [24] would require a substantial reduction of the prefactor. The issue of screening for Fe in Ag is unclear. At first glance, the five channels corresponding to the d-orbitals should overscreen the $s=2$ Fe spin. In reality, however, the different channels have different coupling constants, and the impurity spin and orbital degeneracies are broken by crystal fields and spin-orbit scattering. Further theoretical work on realistic models of spin-2 systems should help to resolve these issues.

Note added: We recently became aware of a related work leading to similar conclusions [25].

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